The Journal of Adhesion, 78: 189–200, 2002
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0021-8464/02 $12.00 +.00

EFFECTS OF THE TEMPERATURE, HUMIDITY, AND STRESS ON THE INTERLAMINAR INTERFACE OF CARBON FIBER POLYMER-MATRIX COMPOSITES, STUDIED BY CONTACT ELECTRICAL RESISTIVITY MEASUREMENT

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The interlaminar interface (i.e., the interface between laminae) of continuous carbon fiber polymer-matrix structural composites was monitored in real time during dynamic changes in temperature, humidity and stress by measurement of the contact electrical resistivity of the interface. The stress was compressive, in the direction perpendicular to the interlaminar interface. Temperature, humidity and stress were all found to have reversible effects on the resistivity, due to the effect of temperature on the probability of the jump of an electron from one lamina to the adjacent one, and the effects of humidity and stress on the extent of contact between fibers of adjacent laminae. In addition, due to damage, temperature caused the resistivity to increase whereas stress caused the resistivity to decrease.

Keywords: Composite; Polymer; Epoxy; Carbon fiber; Electrical resistivity; Humidity

INTRODUCTION

Advanced structural composites are mainly polymer-matrix components containing continuous fibers such as carbon fibers, which are attractive for their high modulus, high strength, low density and thermal conductivity. Among the polymer matrices used for carbon fiber composites, epoxy (a thermoset) is most common.

Received 28 July 2001; in final form 7 December 2001.

This article is one of a collection of papers honoring Hatsuo (Ken) Ishida, the recipient in February 2001 of *The Adhesion Society Award for Excellence in Adhesion Science, sponsored by 3M*.

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A polymer-matrix composite comprising layers (laminae) of continuous fibers tends to be mechanically weakest at the interface between the laminae. As a result, delamination is a common mechanism of failure in the composites. The study of the interlaminar interface has been previously performed by measuring the interlaminar shear strength (ILSS) by techniques such as the short-beam method [1], the Iospiescu method [2] and other methods [3]. Although ILSS is a valuable quantity that describes the mechanical property of the joint between laminae, it gives little information on the interfacial microstructure, such as the extent of direct contact (with essentially no polymer matrix in between) between fibers of adjacent laminae and the residual interlaminar stress resulting from the anisotropy between adjacent laminae. The anisotropy is severe when the fibers in the adjacent laminae lie in different directions, since the fibers and polymer matrix differ greatly in modulus and thermal expansion coefficient. Direct contact between fibers of adjacent laminae occurs due to the flow of the matrix during composite fabrication and the waviness of the fibers. Direct contact means that the thickness of the matrix between the adjacent fibers is so small (say, a few Å) that electrons can tunnel or hop from one fiber to the other. The presence of direct contact has been shown by the fact that the volume electrical resistivity of carbon fiber epoxy-matrix composites in the through-thickness direction is finite, even though the epoxy matrix is electrically insulating [4].

In contrast to other workers, we use the contact electrical resistivity of the interlaminar interface as a quantity to describe the structure of this interface. Note that the volume electrical resistivity is a geometry-independent quantity that describes the resistivity of a three-dimensional material in a particular direction. For example, the volume resistivity of a composite in the through-thickness direction reflects both the volume resistance within each lamina in the through-thickness direction and the contact resistance at each interlaminar interface. Hence, the volume resistivity does not simply relate to the structure of the interlaminar interface. However, the contact resistivity does, since it is a geometry-independent quantity that describes the resistivity of a plane in the direction perpendicular to the plane. The volume resistivity has the unit Ω.cm, whereas the contact resistivity has the unit $\Omega.cm^2$.

For a composite with electrically-conducting fibers, such as carbon fibers, and an electrically-insulating matrix, such as epoxy, the contact resistivity can be conveniently measured, since the fibers serve as electrical leads. The contact resistivity is lower when the extent of direct contact between fibers of adjacent laminae is greater. However, the contact resistivity also depends on the nature of each direct contact. This nature is reflected by the activation energy for electrons to jump from one lamina to an adjacent one. This activation energy is expected to increase when the interlaminar stress is higher. The jumping of the electrons from one lamina to another is a thermally-activated process, so the higher the temperature, the higher is the contact conductivity.

Moisture is known to affect negatively numerous properties of polymers and their composites. Considerable attention has been given by numerous workers to address the effect of moisture on the mechanical behavior of polymer-matrix composites, as the mechanical behavior is relevant to the effectiveness for structural applications. In the case of carbon fiber epoxy-matrix composites, the properties which are dominated by the matrix or the fiber-matrix interface are degraded by moisture absorption, whereas the properties that are dominated by the fibers are essentially not affected [5]. In particular, the interfacial strength [6], the interlaminar tensile strength [7], the mode II critical strain-energy release rate [8], and the mode II interlaminar fracture toughness [9,10] are degraded by moisture. The degradation is attributed to the weakening of the fiber-matrix bond [5, 11], the swelling action of the water [12], the softening of the matrix [5, 11] and the loss of shear strength of the matrix [10]. On the other hand, the curing residual stress is decreased by moisture [7] and the matrix can be plasticized by water [12], thereby increasing the fracture (delamination) toughness [12] or causing moisture to have little effect on the fracture properties [13] in some cases. The moisture effect is aggravated greatly by increasing the temperature [14–17], by using glass fiber in place of carbon fiber [18, 19] or by subjecting the composite to stress [20]. The composite material properties that are affected negatively by moisture include the stiffness [21, 22], the erosion resistance [23], the friction and wear properties [24], the creep compliance [25], the damping ratio [26], the maximum service temperature [27], and the resistance to curvature in the case of non-symmetric laminates [28]. The problem can be alleviated by surface treatment of the carbon fiber [29–31]. The moisture absorption proceeds by diffusion and the absorption is at least partially reversible [32]. In contrast to prior work [5–32], this work uses electrical resistivity measurement to investigate the effect of moisture on carbon fiber epoxy-matrix composites.

Sensing is the most basic function of a smart structure. The sensing of strain, stress, temperature and damage is of particular interest, as strain/stress sensing pertains to structural vibration control and load monitoring, temperature sensing pertains to thermal control, and damage sensing pertains to structural health monitoring.

Strain sensing has been attained in carbon fiber polymer-matrix composites by using the piezoresistive behavior of the bulk composite [33–40]. This behavior involves the volume electrical resistivity of the composite in the longitudinal (fiber) direction decreasing reversibly upon longitudinal tension and that in the through-thickness direction increasing reversibly upon longitudinal tension. The use of the volume resistivity distribution to determine a two-dimensional strain distribution is tedious, as it requires the application of a two-dimensional array of electrical contacts.

In this work, the interlaminar interface is used as a piezoresistive stress (compressive) sensor. The piezoresistivity is associated with the effect of stress on the contact resistivity of the interlaminar interface. By using two crossply laminae, a two-dimensional array of strain sensors and an x-y grid of electrical interconnections are obtained (Figure 1), thus, allowing compressive stress distribution sensing, in which the composite is utilized as both sensors and electrical interconnections.

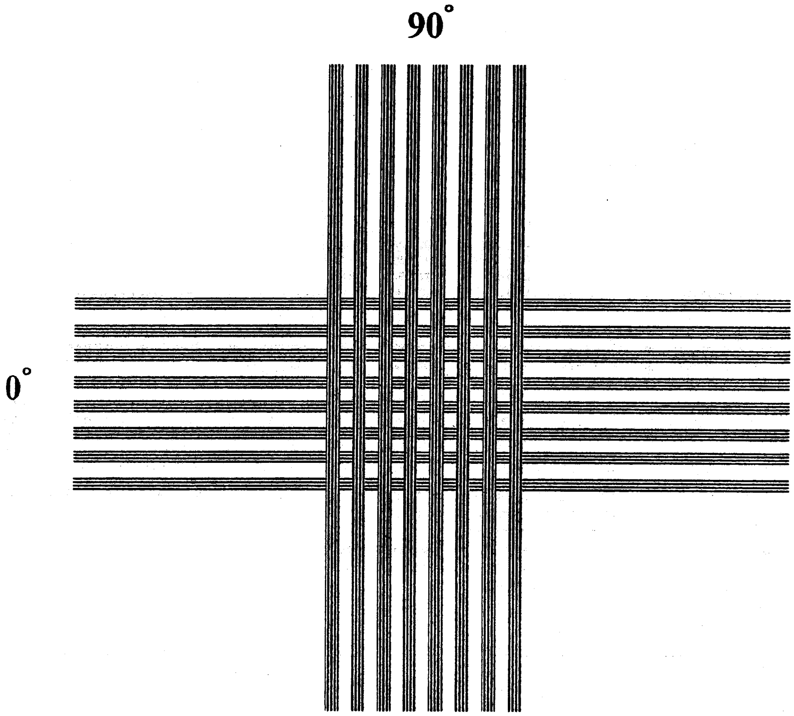

FIGURE 1 Sensor array in the form of a carbon fiber polymer-matrix composite comprising two crossply laminae.

EXPERIMENTAL METHODS

Two laminae of unidirectional carbon fiber epoxy-matrix prepregs (provided by Cape Composites Inc., San Diego, CA, USA) (Table 1) in the form of strips crossing one another, with one strip on top of the other (Figure 2), were fabricated into a composite at the overlapping region (ranging from 3 × 3 to 6 × 6 mm) of the two laminae by applying pressure and heat to the overlapping region (without a mold). The pressure was provided by weights. A glass fiber epoxy-matrix composite spacer was placed between the weight and the junction (the overlapping area region of the two strips). The heat was provided by a Carver hot press. A Watlow model 981C-10CA-ARRR temperature controller was used to control the temperature and the ramping rate. Each of the specimens was put between the two heating platens of the hot press and heated linearly up to $121 \pm 2°C$ at the rate of 2°C/min. Then it was cured at that temperature for 3 h and subsequently furnace cooled to room temperature.

Thermal cycling was conducted after curing and subsequent cooling of the composite by using a small resistance heater for heating and using compressed air and copper tubing with flowing water for cooling. All the time, the contact electrical resistance and the temperature of the sample were measured, respectively, by a Keithley (Keithley Instruments, Inc., Cleveland, OH, USA) 2001 multimeter and a T-type thermocouple, which was put just beside the junction. Electrical contacts were made to the four ends of the two strips, so as to measure the contact electrical resistivity (resistance multiplied by contact area, which is the area of the overlapping region) between the two laminae in the composite, using the four-probe method (Figure 2). The epoxy at the ends of each prepreg strip was burnt out to expose the carbon

TABLE 1 Carbon Fiber and Epoxy Matrix Properties (According to Cape Composite Inc., San Diego, CA, USA)

Fortafil 555 continuous carbon fiber	
Diameter	6.2 μm
Density	1.8 g/cm^3
Tensile modulus	231 GPa
Tensile strength	3.80 GPa
Cape C2002 epoxy	
Processing temperature	121°C
Flexural modulus	99.9 GPa
Flexural strength	1.17 GPa
T_g	129°C
Density	1.15 g/cm^3

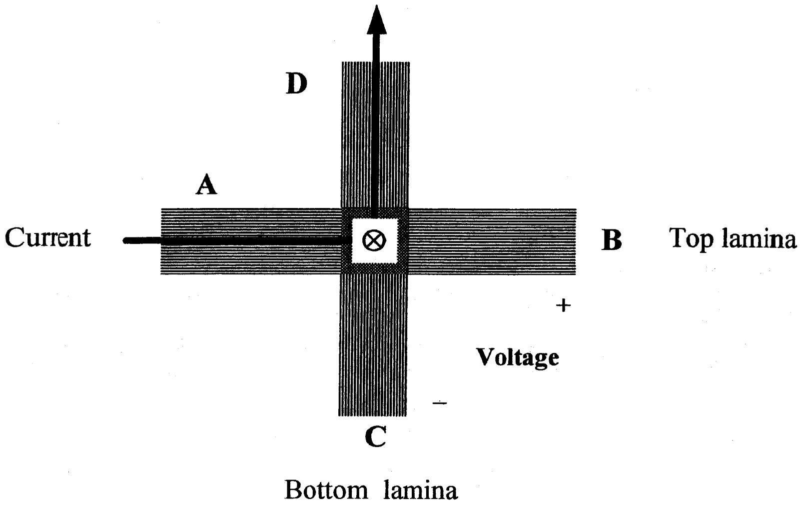

FIGURE 2 Composite configuration for measuring the contact electrical resistivity.

fibers for the purpose of making electrical contacts. These exposed fibers were wrapped by pieces of copper foil, with silver paint between the copper foil and the fibers. The electric current flowed from A to D, such that the dominant resistance was the contact resistance as the volume resistance of the strips was negligible in comparison. The voltage between B and C is the voltage between the two laminae.

Humidity variation was conducted after curing and subsequent cooling of the composite by using liquid water as the source of water vapor. All the time, the contact electrical resistance and the relative humidity were measured, respectively, by a Keithley 2001 multimeter and a humidity sensor (Honeywell Micro Switch, Morristown, NJ, USA HIH-3605-A-CP).

A dynamic compressive stress was applied on the overlapping region (Figure 2) by using a hydraulic mechanical testing system (MTS 810, MTS Systems Corp., Marblehead, MA, USA). Simultaneously, the contact electrical resistance was measured.

RESULTS AND DISCUSSION

As shown in our earlier work, the initial contact resistivity (*i.e.*, before variation of temperature, humidity or stress) decreased with increasing curing pressure during composite fabrication [41]. The differences in the initial contact resistivity among the specimens in this work are primarily due to the differences in curing pressure.

Figure 3 shows the variation of the contact resistivity with temperature during thermal cycling for a composite fabricated at a curing pressure of 0.33 MPa. The temperature was repeatedly increased to

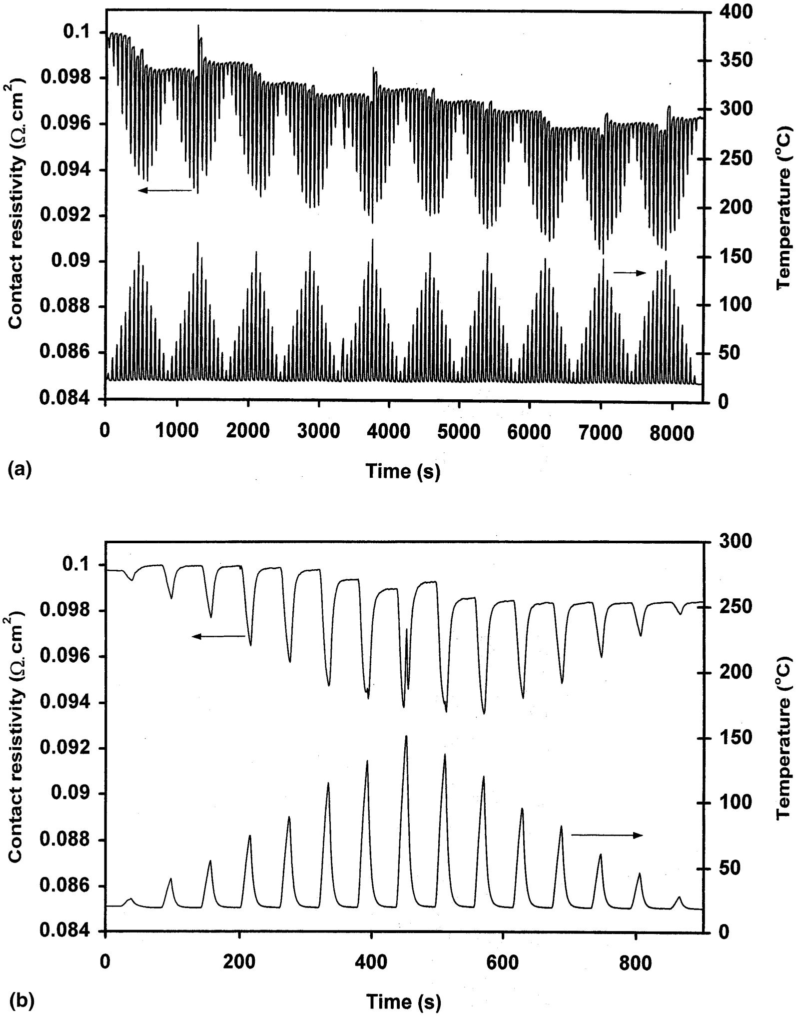

FIGURE 3 Variation of the contact electrical resistivity with time and of the temperature with time during thermal cycling. (a) The first 10 groups. (b) The first group.

various levels. A group of cycles in which the temperature amplitude increased cycle by cycle and then decreased cycle by cycle back to the initial low temperature amplitude is hereby referred to as a group. The contact resistivity decreased reversibly upon heating in every cycle of every group, as shown in Figure 3(a) for the first 10 groups and in Figure 3(b) for the first group. The higher the temperature, the lower was the contact resistivity. At the highest temperature (150°C) of a group, a spike of resistivity increase occurred, as shown in Figure 3(b). This spike was observed similarly in other groups. It is attributed to damage at the interlaminar interface.

Figure 4 shows the variation of the contact resistivity with time and of the relative humidity with time during cycling of the relative humidity for the composite made at a curing pressure of 0.21 MPa. The resistivity decreased reversibly upon humidity increase. The reversibility is essentially complete after the first cycle of humidity variation. This trend is attributed to the distance between the fibers of adjacent laminae increasing as the epoxy matrix between the laminae expands upon moisture uptake.

Moisture causes expansion of the epoxy matrix, as discussed above. On the other hand, an increase in temperature also causes expansion

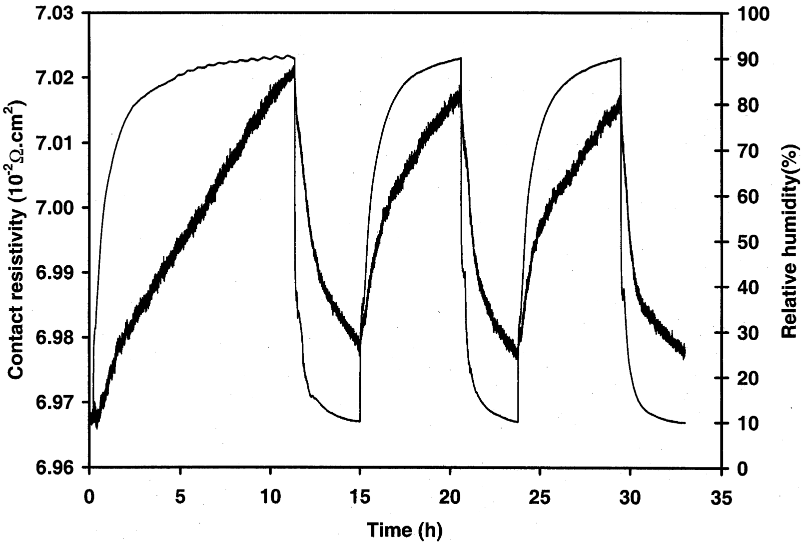

FIGURE 4 Variation of the contact electrical resistivity (thick curve) with time and of the relative humidity (thin curve) with time during humidity variation for composite made at a curing pressure of 0.21 MPa.

of the epoxy matrix, due to thermal expansion. In our previous study of the effect of temperature on the contact resistivity, we observed that an increase in temperature caused the resistivity to decrease, irrespective of the curing pressure [41]. This suggests that the expansion resulting from moisture uptake is not the same as that resulting from heating. The relief of residual stress upon heating is significant, whether the curing pressure is high or low.

Figure 5 shows the variation of the contact resistivity with stress during compressive stress cycling to various maximum stresses up to 4 MPa. The composite was made at a curing pressure of 0.43 MPa. The contact resistivity decreased quite reversibly upon loading, due to the increased contact between fibers of adjacent laminae. However, the resistivity decrease was not completely reversible. The greater the stress, the more the contact resistivity decreased. Although Figure 5 shows results at stress amplitudes up to 4 MPa, similar results were obtained up to 26 MPa.

The upper envelope of the resistivity variation in Figure 5 decreased gradually cycle by cycle. This means that the resistivity decrease upon loading was not totally reversible. The partial irreversibility means that the increase in the extent of contact between fibers of adjacent laminae upon loading is not completely reversible.

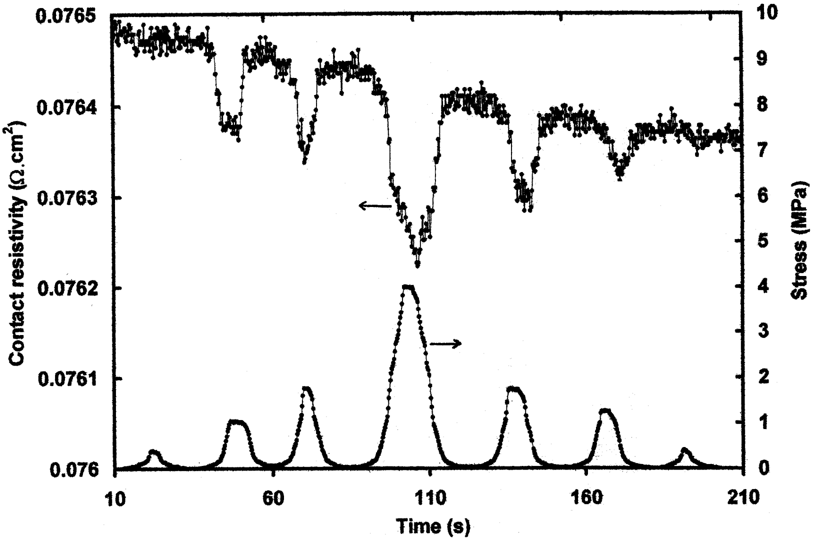

FIGURE 5 Variation of the contact electrical resistivity with time and of the stress with time during stress cycling to different stress amplitudes up to 4 MPa.

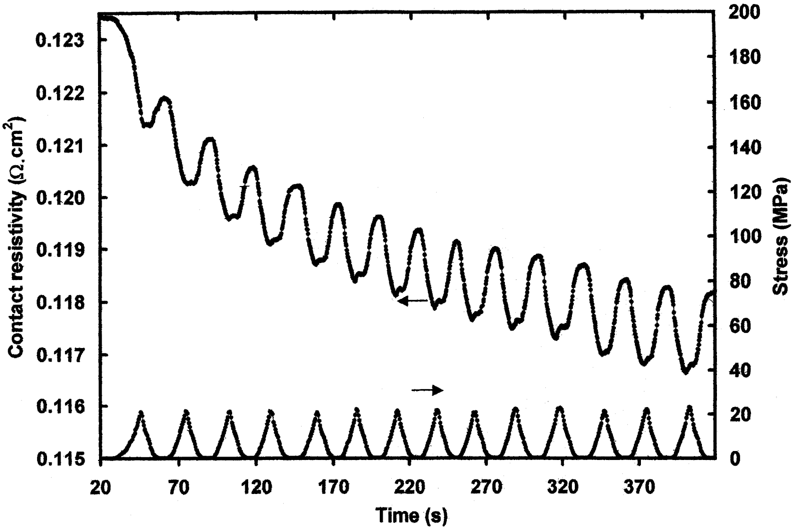

FIGURE 6 Variation of the contact resistivity with time and of the stress with time during stress cycling at a constant stress amplitude of 20 MPa.

Stress cycling at a fixed stress amplitude of 20 MPa for 14 cycles (Figure 6) showed that both the upper and lower envelopes of the resistivity decreased irreversibly and gradually leveled off as cycling progressed, while the reversible effect within a cycle was essentially not affected. The irreversible effect is a form of minor damage of the interlaminar interface. The damage was most significant in the first two cycles and subsequent incremental damage diminished as cycling progressed.

CONCLUSION

Temperature, humidity and stress (compressive, perpendicular to the laminae) have reversible effects on the contact electrical resistivity of the interlaminar interface of a crossply carbon fiber epoxy-matrix composite. Temperature affects the probability of the jump of an electron from one lamina to the adjacent one. Thus, the higher the temperature, the lower is the contact resistivity. Humidity and stress affect the extent of contact between fibers of adjacent laminae. The higher the stress, the lower is the contact resistivity. An increase in humidity causes the resistivity to increase reversibly. Thermal damage causes the resistivity to increase, whereas compressive stress damage causes the resistivity to decrease. The reversible effects allow the use of the contact resistivity as an indicator of temperature, humidity and stress. The irreversible effects allow structural health monitoring.

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